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(54) Title: EXTRACTION OF MATERIALS FROM PLANTS

(57) Abstract: Method for extracting materials, particularly bio-active materials having insecticidal, bactericidal and/or insect repellent properties from plants such as those of the Chrysanthemum family or of the *Helianthus* family, by solvent extraction using plant-derived solvent such as terpenes e.g. terpineol and plant oils.

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WO 01/07135 A2

(54) Title: EXTRACTION OF MATERIALS FROM PLANTS

(57) Abstract: Method for extracting materials, particularly bio-active materials having insecticidal, bactericidal and/or insect repellent properties from plants such as those of the Chrysanthemum family or of the *Helianthus* family, by solvent extraction using plant-derived solvent such as terpenes e.g. terpineol and plant oils.

EXTRACTION OF MATERIALS FROM PLANTS

Field of the Invention

This invention concerns the extraction of materials from plants, particularly bio-active materials, that is materials having insecticidal, bactericidal and/or insect repellent properties.

Background to the Invention

It is known to extract bio-active material known as pyrethrum from flowers of the plant *Chrysanthemum cinerariaefolium*. Pyrethrum is a mixture of substances that acts as a powerful but non-persistent contact insecticide with rapid "knockdown" effect, while being non-toxic to mammals.

The normal methods of extraction of pyrethrum are complex, multi-step processes that involve heating of organic solvents derived from petroleum, and require several stages of concentration, selective separation and refinement which are carried out using expensive equipment. A typical pyrethrum extraction process involves the following steps:

1. Flowers are gathered at full bloom
2. Flowers are dried, usually in the open air.
3. Dried flowers are transported to a central processing facility.
4. Dried flowers are ground to fine powder.

5. Ground flowers are extracted using hexane solvent at elevated temperature.

6. Hexane solvent is distilled off leaving a dark tarry oleoresin.

7. Oleoresin is refined by further selective solvent extraction performed at elevated temperature to provide a light coloured liquid.

8. Refined extract is diluted using mineral oil to form a 25% (by weight) solution ready for sale and use.

The process produces approximately 3% by weight pure pyrethrum extract from dry flowers.

The present invention is based on a novel method of extracting materials from plants. The present invention further concerns extraction of bio-active material and associated material from the order *compositi*, in particular of plants in the *Chrysanthemum* family and of plants in the *Helianthus* family commonly known as Sunflowers, in particular from *Helianthus Annuus* which is cultivated in many countries for the production of Sunflower seed oil. A feature of the Invention is that it uses the Sunflower head after removal of the seeds and this part of the flower is of no value and is discarded as waste.

Summary of the Invention

In a first aspect, the invention provides a method of extracting materials from plants, comprising mixing plant matter with one or more solvents derivable from plants, resulting in extraction into the solvent or solvents of material from the plant matter.

The plant matter conveniently comprises flowers or flower heads (without petals), which may be in fresh or dried condition.

The plant matter may be reduced to smaller pieces for processing, e.g. by cutting, shredding or grinding.

The solvent or solvent mixture conveniently comprises one or more terpenes (including sesquiterpenes, diterpenes and higher polymers and various oxygen-containing compounds from terpenes, such as alcohols, eg terpineol, ketones and camphors) and plant oils e.g. rosemary oil and lavender oil. Currently preferred plant solvents include the terpenes, terpineol, D-limonene, α-pinene, β-pinene and mixtures thereof, e.g. a mixture of equal parts by weight of terpineol and D-limonene.

The mixture is preferably agitated or stirred, desirably in a way that reduces the size of pieces of the plant matter to expose a greater surface area of plant matter to the solvent and increase the speed of the extraction process. This is conveniently achieved using a mechanical macerating mixer. Extraction starts to occur relatively rapidly (within a few

minutes) even at ambient temperature (about 20°C) and occurs more rapidly with heating, and is typically complete within about 10 minutes even at ambient temperature. Heating, however, is not currently favoured as this may cause evaporation of one or more bio-active ingredients of the material, reducing efficacy of the product.

After extraction, the residual solid plant matter is preferably removed, e.g. by filtration, leaving solvent having dissolved therein extracted plant material.

Alternatively, the solvent may be removed or reduced in amount, e.g. by evaporation or distillation.

The method is applicable to a range of plants, particularly those containing extractable bio-active materials having insecticidal, bactericidal and/or insect repellent properties. In particular the method is applicable to plants of the order *compositi*, in particular the *Chrysanthemum* family, such as marigold plants and also *Leucanthemum vulgare* (commonly known as the Oxeye daisy), and the *Helianthus* family, such as *Helianthus Annuus* San Luca.

The materials extracted by the method of the invention can find use as bio-active materials, particularly as insecticides, bactericides and/or insect repellents. Therefore, in a further aspect, the present invention provides a composition comprising extracted plant material capable of substantially repelling or killing insects, and/or killing micro-organisms.

It has been found that an extract solution obtained from 20 grams of dried Sunflower extracted with 100 grams of a mixture of 80 grams of D-Limonene and 20 grams of Terpineol is insecticidal as prepared. Thus, in a further aspect, the present invention provides a mixture of the extract in D-Limonene or in D-Limonene and Terpineol, for example. The inclusion of Terpineol in the mixture can provide fungus and mould resistance.

In yet a further aspect the invention covers material extracted by the method of the invention.

It has also been found that a solution in D-Limonene, when allowed to evaporate to remove the D-Limonene leaves a residue of extracted material which is both repellent and insecticidal to flies.

Another aspect of the invention thus provides a composition comprising material extracted by the method of the invention.

In a simple case, the composition may comprise extracted plant material in the solvent or solvents used for the extraction process, as discussed above. As the solvent is plant-derived, the composition may be fully plant-derived. The solvent or solvents may be used as a carrier for the bio-active material when formulated into a product and may be selected having regard to the intended use of the composition, to impart desired properties to the composition such as odour, evaporation rate and other physical and chemical properties.

Such a composition may be used as it is as an insecticide, bactericide and/or insect repellent.

The composition need not include solvent used for extraction purposes. Such solvent may be removed, e.g. by evaporation or distillation.

The composition may include optional additional ingredients selected having regard to the intended use of the composition. For example, a composition intended for use as an insecticidal or insect repellent cream, lotion, ointment, spray, etc may include a base or vehicle known to those skilled in the art for such purposes.

In yet a further aspect, the invention provides material obtainable from a plant from the order *compositi*, in particular, a member of the *Chrysanthemum* family, such *Leucanthemum vulgare*, or a member of the *Helianthus* family, such as *Helianthus annuus* San Luca.

The plant *Leucanthemum vulgare* is commonly known as the Oxeye daisy. This plant grows widely in the wild in the United Kingdom and elsewhere, and is commonly regarded as a weed.

The plant *Helianthus annuus* is commonly known as Sunflowers, and is cultivated in many countries for the production of Sunflower seed oil.

The material is preferably obtained from plant matter comprising flowers or flower heads (without petals), which may be in fresh or dried condition. The plant matter may be reduced to smaller pieces for processing, e.g. by cutting,

shredding or grinding.

The extracted material can be used as an ingredient of a composition, particularly a composition having insecticidal, bactericidal and/or insect repellent properties. The extracted material has been found to be bio-active and to having insecticidal, bactericidal and/or insect repellent properties. These properties are observed on contact with or in proximity to the material and/or vapour therefrom. Not wishing to be bound by theory, it is thought that the material is not pyrethrum-like in nature but instead may comprise a mixture of terpenes. The material appears to have stronger insecticidal properties than pyrethrum alone. Moreover, the material may be easily produced using simple extraction techniques not requiring heating and the extracted active material has greater efficacy and better retention than pyrethrum from the known extraction processes.

In a further aspect the invention thus provides a composition comprising material obtainable from a plant the order *compositi*, in particular, a member of the *Chrysanthemum* family, such *Leucanthemum vulgare*, or a member of the *Helianthus* family, such as *Helianthus annuus* using the above described method of the first aspect of the present invention.

In another aspect of the invention provides a method of killing or repelling insects or killing microorganisms, comprising exposure to material extractable from the plant *Leucanthemum vulgare* or *Helianthus annuus* San Luca.

A further aspect of the invention resides in a method of producing material having insecticidal, bactericidal and/or insect repellent properties, comprising extracting material from a plant from the order *compositi*, in particular, a member of the *Chrysanthemum* family, such *Leucanthemum vulgare*, or a member of the *Helianthus* family, such as *Helianthus annuus* San Luca.

Extraction is preferably performed by solvent extraction, as described above.

The method of the invention can be performed simply without the need for complex, costly equipment, and can be performed rapidly and efficiently at ambient temperature (about 20°C). This thus provides benefits and advantages compared with the conventional method for extracting pyrethrum, as discussed above, which are complex, multi-step processes involving several high temperature steps.

Furthermore, an emulsion of the solvent and water extracts in water can form a material suitable for spray application as an insecticide against flying and crawling insects. This mixture can form a material suitable for spray application on agricultural and horticultural plants to protect from insect pests and fungus with the additional benefit of the material being biodegradable.

The invention will be further described, by way of illustration, in the following Examples with References to the accompanying Figures, in which:

Figure 1 illustrates a graph depicting the effect of material extracted from *Leucanthemum vulgare* on flies.

In the Examples, all percentages are by weight unless otherwise stated.

Example 1

Material was extracted from the plant *Leucanthemum vulgare* in the following way.

Flowers of the plant *Leucanthemum vulgare* were gathered at full bloom or harvested at petal drop and dried in conventional manner. The dried flowers were mixed with terpineol (20 grams of dried flowers to 100 grams of terpineol) and the mixture ground at ambient temperature (about 20°C) in a high speed mechanical macerating mixer with cutting head in the form of a Silverson laboratory mixer (Silverson is a Trade Mark), which acts to cut the dried flowers into small fragments and mix them intimately with the terpineol. Within a few minutes the colour of the terpineol starts to change, becoming yellow, as a result of solvent extraction of material from the flowers. Mixing is continued until no further colour change in the terpineol is observed, i.e. the terpineol acquires a maximum density of yellow colour, indicating maximum extraction. This occurs after about 10 minutes. The mixer is switched off, and the resulting mixture is filtered in conventional manner to remove ground flower residue, leaving a yellow liquid comprising terpineol

with dissolved material extracted from *Leucanthemum vulgare*.

The separated ground flower residue is rinsed with a fresh batch of terpineol to remove remaining material from the residue, and this batch of terpineol is used for extraction of the next batch of flowers.

The yellow liquid produced by treatment of the first batch of flowers as described above has bio-active properties and may be used in this form, without further treatment, on its own or as an ingredient in a composition, e.g. an insecticidal cream, where it will exhibit bio-active properties as described above. Alternatively, the yellow liquid may be treated to remove some or all of the terpineol, e.g. by heat treatment or distillation, to produce a more concentrated product. This product can again be used on its own or as an ingredient in a composition.

The process produces pure extracted bio-active material from dry flowers in an amount of about 5% by weight of the flowers.

Example 2

Material produced as described in Example 1, in the form of the yellow liquid of maximum density of yellow colour, was tested for insecticidal properties. About 1 grams of the liquid was placed on filter paper and allowed to age for several hours. The filter paper was then placed in a container with live flies. All of the flies died within about 4 minutes.

By way of comparison, terpineol alone used in the same way under the same conditions did not kill flies.

Example 3

Material was produced as described in Example 1, but using D-limonene or a mixture of equal parts by weight of D-limonene and terpineol in place of terpineol. The extracted material, in the form of the yellow liquid of maximum density of yellow colour, was used as an ingredient in an insect repellent product intended for use on the skin as protection against mosquitoes and other flying insects. The insect repellent product formulation was as follow:

Extract	3.0 grams
Water	47.0 grams
Corn Oil	47.0 grams
Alginate (seaweed extract)	2.0 grams
Non-ionic surfactant	1.0 grams

Example 4

Material was produced as described in Example 1, but using D-limonene in place of terpineol. The extracted material, in the form of the yellow liquid of maximum density of yellow colour, was concentrated by evaporation of the D-limonene until no solvent remains. The vapour produced from the remaining extracted material (which is in the form of an

essential oil) has bactericidal properties, e.g. against methicillin resistant *Staphylococcus aureus*, *Burkholderia cepacia* and *Escherichia coli*. Tests were carried out in a Petri dish at an appropriate temperature for bacterial culture (about 37°C).

Example 5

A comparison was made of the insecticidal efficacy of pyrethrum (in the form of a 25% solution in mineral oil) extracted in conventional manner and material extracted from *Leucanthemum vulgare* by the method of the invention.

The materials tested were as follows:

1. 25% pyrethrum solution obtained from The Pyrethrum Board of Kenya.
2. 20% solution of material prepared by the method in accordance with the invention, prepared from dried mature flowers of *Leucanthemum vulgare* as described in Example 1 by processing a mixture of 20 grams flower to 80 grams terpineol using a Silverson mixer.
3. 20% solution prepared as described in 2 above but using a serrated head stirrer (having a 64mm diameter serrated disc, rotating at 1000 rpm) in place of a Silverson mixer.

Equal volumes (about 0.5 grams) of the above 3 materials were placed in the centre of respective Whatman No. 1 filter papers 11cm in diameter and allowed to settle for about 1 hour uncovered at ambient temperature (about 20°C). The resulting

colours on the filter papers were as follows:

1. medium red/brown

2. stronger red/brown

3. very little colour, indicating less effective extraction of active material.

No significant chromatographic separation of any of the samples on the filter papers occurred during this time.

Efficacy tests were carried out by placing a respective inverted glass jar or beaker over each - filter paper and placing two live flies of similar type in each container at various different times after the start of the tests. The tests were started about 1 hour after placing of samples on the filter papers. The effect of the materials on the insects with time was observed, with the time (in minutes) taken for the flies to die or become moribund being noted. Test results for the three different materials, using the same identification numbers as above, were as follows:

Time of day	Material 1	Material 2	Material 3
11am	7	2	5
Noon	7	2	no effect
2pm	3	3	no effect
4pm	5	5	no effect
2am (next day)	10	12	no effect

The following remarks and observations are offered.

1. Sample 2 (prepared in accordance with the invention) showed more effective results compared with sample 1 (pyrethrum) at early stages of the tests. At later stages these two samples gave similar results.

2. The early higher efficacy of sample 2 as compared with sample 1 indicates the presence of a more volatile bio-active component in sample 2 as compared with sample 1.

3. Sample 3 gave poor results after 1 hour from start of the tests. This indicates that sample 3 contains mainly terpineol and very little extracted active material. This indicates that the solvent component of samples 2 and 3 is not effective against the flies, and that also it is preferred to use a Silverson mixer for extraction.

4. During the test period the colour of samples 1 and 2 gradually faded. At the end of the test sample 2 was pale yellow and sample 1 was a darker yellow/brown.

Example 6

Helianthus Annuus San Luca extracted in D-limonene in accordance with Example 1 was allowed to evaporate at room temperature and residue collected from open trays. A smear of residue approx. 2.5cm in diameter was made on aluminium foil. Flies were collected and placed in an upturned glass container. Several different species of flies were tested including black flies, midges and a type of wasp.

All flies were immediately affected and showed signs of distress. When forced to come into contact with the smear, all became moribund within 15 minutes and death resulted after 1 hour.

These results indicate that the residue is repellant in the vapour phase to the flies tested and is toxic through contact, indicating a strong insecticide action.

Example 7

Extracts from *Leucanthemum vulgare* in D-limonene were allowed to evaporate at room temperature and residue collected from open trays. A smear of residue approx. 2.5cm in diameter was made on Whatman filter paper. This was also performed for Pyrethrum. Flies were collected and placed in an upturned glass container. Several different species of flies were tested including black flies, midges and a type of wasp.

Flies were forced to contact the residues and were killed within 2 minutes. Flies were replenished at timed intervals and the length of time taken to kill them measured. With pyrethrum, the time increased rapidly within 24 hours, so that it took more than 9 minutes to kill the flies. The extract from *Leucanthemum vulgare*, however, maintained its strength until after 96 hours. It was not until 200 hours after application that the *Leucanthemum vulgare* extract took as long to kill as pyrethrum after 24 hours.

CLAIMS

1. A method of extracting materials from plants, comprising mixing plant matter with one or more solvents derivable from plants, resulting in extraction into the solvent or solvents of material from the plant matter.

2. A method according to claim 1 wherein said plants belong to a family selected from the group comprising *Chrysanthemum* and *Helianthus*.

3. A method according to claim 2 wherein said plant is *Leucanthemum vulgare*.

4. A method according to claim 2 wherein said plant is *Helianthus annuus*.

5. A method according to claims 2, wherein said material is extracted from plant matter comprising flowers or flower heads, in fresh or dried condition from a plant selected from the group comprising *Leucanthemum vulgare* and *Helianthus annuus*.

6. A method according to any one of claims 1, 2, or 5 wherein said material is extracted by solvent extraction.

7. A method according to claim 6, wherein said solvent extraction is carried out using solvents derivable from plants.

8. A method according to claim 6 wherein said solvent comprises one or more terpenes and/or plant oils.

9. A method according to claim 8 wherein said solvent comprises terpineol and/or D-limonene.

10. A method according to claim 1, 2 or 5, wherein said plant matter is reduced to smaller pieces for processing.

11. A method according to any one of the preceding claims, wherein said solvent extraction is carried out at ambient temperature.

12. A method according to any one of the preceding claims wherein said solvent extraction is carried out in a mechanical macerating mixer.

13. A method according to any one of the preceding claims wherein after said extraction is performed, residual solid plant matter is removed, leaving solvent having dissolved therein extracted plant material.

14. A method according to claim 13 wherein after removal of residual solid plant matter, solvent is removed or reduced in amount.

15. A method according to any one of claims 1 to 14 wherein the material produced has insecticidal, bactericidal and/or insect repellent properties.

16. Insecticidal, bactericidal and/or insect repellant material extracted from a plant by the method of any one of the preceding claims.

17. Material from a plant belonging to the order *compositi*, wherein said material is obtainable by extracting the material from plants using one or more solvents derivable from plants.

18. Material according to claim 17 wherein said material is obtainable from a plant belonging to a family selected from the group comprising *Chrysanthemum* and *Helianthus*.

19. Material according to claim 18 wherein said material is obtainable from the plant *Leucanthemum vulgare*.

20. Material according to claim 18 wherein said material is obtainable from the plant *Helianthus annuus*.

21. Material according to any one of claims 17 to 20 wherein said material is obtainable from plant matter comprising flowers or flower heads, in fresh or dried condition.

22. Material according to any one of claim 17 to 21 wherein said material is obtainable by solvent extraction.

23. Material according to claim 22 wherein said solvent extraction is carried out using one or more solvents derivable from plants.

24. Material according to claim 23 wherein said solvent comprises one or more terpenes or plant oils.

25. Material according to claims 22, 23 or 24, wherein said solvent extraction is performed at ambient temperature.

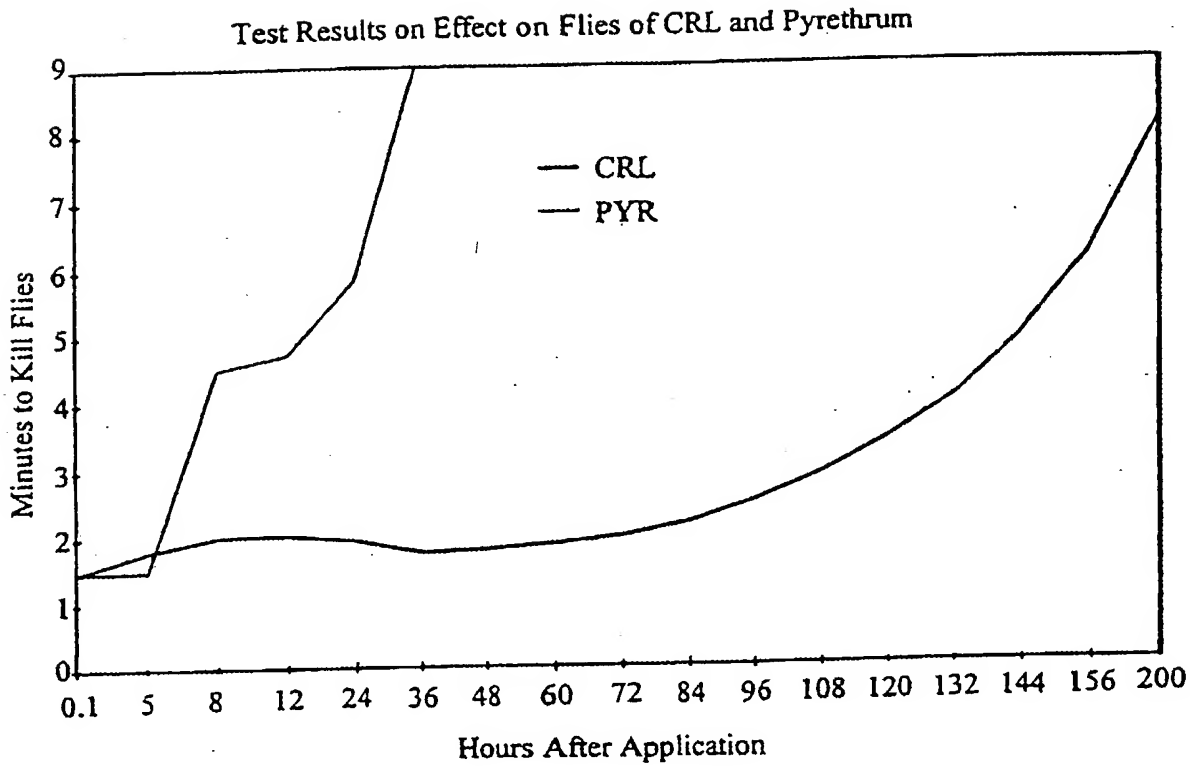
26. A insecticidal, bactericidal and/or insect repellant composition comprising material in accordance with any one of claims 17 to 25 and a carrier.

27. A composition according to claim 26 wherein said composition further comprises one or more solvents used for extraction of the material.

28. A composition according to claim 27 wherein said one or more solvents are plant-derived.

29. A composition according to claims 26, 27 or 28 in the form of an insecticidal, bactericidal or insect repellent composition.

30. A method of killing or repelling insects or killing micro-organisms, said method comprising exposing said insects or micro-organisms to a material or composition in accordance with any one of claims 16 to 29.


Test Results on Effect on Flies of CRL and Pyrethrum
Minutes to Kill Flies
0
1
2
3
4
5
6
7
8
9
CRL
PYR
0.1
5
8
12
24
36
48
60
72
84
96
108
120
132
144
156
200
Hours After Application

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/GB 00/02870

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7. B01D11/02 A01N65/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B01D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR 2 448 856 A (SAPHYR SARL) 12 September 1980 (1980-09-12) claims 1,6-9	1,6-17
A	DATABASE WPI Week 199148 Derwent Publications Ltd., London, GB; AN 1991-346882 XP002154928 & CN 1 050 124 A (W.QINGREN), 27 March 1991 (1991-03-27) abstract	2,18
A	US 5 411 736 A (J.C.LOCKE ET AL.) 2 May 1995 (1995-05-02) claim 1	1

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
19 December 2000	11/01/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Bertram, H

INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 350 897 A (STANDARD OIL) 15 June 1931 (1931-06-15) page 2, line 37 - line 54; claim 1	1

INTERNATIONAL SEARCH REPORT

...formation on patent family members

Inter...nal Application No
PCT/GB 00/02870

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
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